Experimental violation of a spin-1 Bell inequality using maximally-entangled four-photon states

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We demonstrate the first experimental violation of a spin-1 Bell inequality. The spin-1 inequality is a calculation based on the Clauser, Horne, Shimony and Holt formalism. For entangled spin-1 particles the maximum quantum mechanical prediction is 2.552 as opposed to a maximum of 2, predicted using local hidden variables. We obtained an experimental value of 2.27 $\pm$0.02 using the four-photon state generated by pulsed, type-II, stimulated parametric down-conversion. This is a violation of the spin-1 Bell inequality by more than 13 standard deviations.

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In 1935 Einstein, Podolsky and Rosen (EPR) showed that quantum mechanics implied nonlocality [1]. In 1951, Bohm discussed correlations of two entangled spin-1/2 particles of the form $\frac{1}{\sqrt{2}}\left(|+\frac{1}{2},-\frac{1}{2}\rangle - |-\frac{1}{2},+\frac{1}{2}\rangle\right)$. A graphical representation of entangled spin-1/2 Bohm-type apparatus is shown in Fig. 1 a). One spin-1/2 particle is sent to Alice who analyzes the particle in a basis determined by her analyzer orientation α. The conjugate particle is sent to Bob who analyzes his particle in a basis determined by his analyzer orientation β. Alice and Bob then measure either $+1/2$ or $-1/2$. Quantum mechanics predicts that Alice's and Bob's measurements are correlated such that they appear to violate EPR's notion of locality. In 1965, Bell [3] and later Clauser, Horne, Shimony and Holt (CHSH) [4] used the spin-1/2 Bohm-type ideas to show that quantum predictions and local explanations of the correlations were mathematically incompatible. Since that time spin-1/2 experiments employing polarization entangled photons [5, 6, 7, 8], momentum entangled photons [9, 10] and most recently with trapped ions [11] have been used to verify the quantum predictions. The nonlocality of these entangled spin-1/2 particles has been employed in several important applications in the field of quantum information such as dense coding [12], quantum cryptography [13, 14] and quantum teleportation [15].

A natural extension of the research on entangled particles is the study of entangled states of spin-s objects ($s > 1/2$). Gisin and Peres showed that entangled particles with arbitrary large spins still violated a Bell inequality[16]. This result implies that large quantum numbers are no guarantee of classical behavior. Apart from its the fundamental interest [16, 17, 18], entangled states of spin-s objects are of clear interest for applications in quantum information due to the higher dimensional Hilbert space associated to these states (e.g. quantum cryptography, dense coding and bound entanglement[19]). Despite the strong interest expressed in entangled spin-s objects no experimental realization has been reported to date. Here we present the first experimental demonstration of a violation of the Bell inequality for entangled spin-1 objects. We use the fact that the polarization entangled four-photon fields (2-photons in each of two spatial modes) of pulsed parametric down-conversion are formally equivalent to two maximally entangled spin-1 particles [20]. This is related to theoretical work by Drummond [21] in which he describes cooperative emission of wavepackets containing N-bosons and proved that multiparticle states could violate the Bell inequalities. The connection between states produced in parametric down-conversion and the N-boson multiparticle states has recently been discussed by Reid *et al* [22].

A spin-1 particle would have three distinct measurement states ($|1\rangle$,$|0\rangle$, or $|-1\rangle$) The spin-1 analog of Bohm's entangled spin-1/2 particles would be given by

$$|\Psi_1\rangle = \frac{1}{\sqrt{3}}\left(|1,-1\rangle - |0,0\rangle + |-1,1\rangle\right) \tag{1}$$

A graphical representation of Bohm-type entangled spin-1 particles is shown in Fig. 1 b). Similar to the spin-1/2 case, one spin-1 particle is sent to Alice who analyzes the particle in a basis determined by her analyzer orientation α. The conjugate particle is sent to Bob who analyzes his particle in a basis determined by his analyzer orientation β.

The crux of Bell inequalities is that the probabilities P of the "locally explicable" outcomes can be decoupled as

$$P(A,B|\alpha,\beta) = P(A|\alpha,\lambda)P(B|\beta,\lambda) \tag{2}$$

where λ accounts for all local hidden variables. A and B refer to the measurement results ($|1\rangle$,$|0\rangle$, or$|-1\rangle$) obtained by Alice and Bob using analyzer orientations α and β respectively. Taking a local realists point of view we assume that the measurement outcomes can be decoupled. We define a spin-1 local hidden variable measurement combination

$$E^{HV}(\alpha,\beta) = \int d\lambda f(\lambda)\overline{A}(\alpha,\lambda)\overline{B}(\beta,\lambda). \tag{3}$$

where

$$\overline{A}(\alpha,\lambda) = P(1|\alpha,\lambda) - P(0|\alpha,\lambda) + P(-1|\alpha,\lambda) \tag{4}$$

$$\overline{B}(\beta,\lambda) = P(1|\beta,\lambda) - P(0|\beta,\lambda) + P(-1|\beta,\lambda) \tag{5}$$

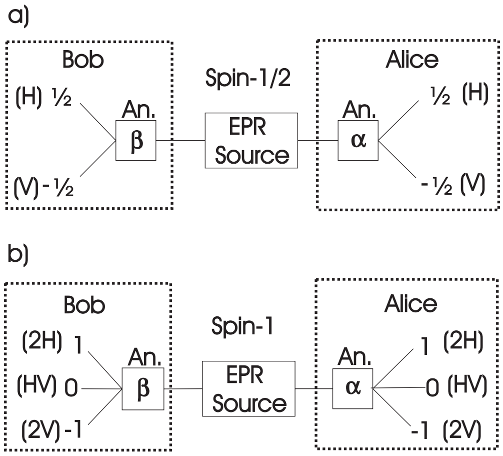

FIG. 1: (a) A Bohm-type spin-1/2 Gedanken experiment. Alice and Bob each receive one particle from an entangled pair. Both Alice and Bob measure either $+1/2$ or $-1/2$ for any orientation of their analyzers (α and β) and their outcomes will be correlated according to quantum mechanical predictions. (b) Bohm-type spin-1 Gedanken experiment. Similarly Alice and Bob measure correlated values of 1, 0 and -1.

Because the signs of the probabilities in both $\overline{A}(\alpha,\lambda)$ and $\overline{B}(\beta,\lambda)$ are different, it must be true that $|\overline{A}(\alpha,\lambda)| \leq 1$ and $|\overline{B}(\beta,\lambda)| \leq 1$. The derivation of the spin-1 Bell inequality proceeds exactly as the spin-1/2 formalism [4, 23], leading to

$$S = |E(\alpha,\beta) - E(\alpha,\beta') + E(\alpha',\beta) + E(\alpha',\beta')| \leq 2 \quad (6)$$

Hence, the maximum possible value that can be achieved, assuming locally explicable outcomes is 2. On the other hand, quantum mechanics states that the measurement probabilities on Alice's and Bob's side cannot be decoupled, which implies that the quantum mechanical measurement outcome is

$$\begin{aligned} E^{QM}(\alpha,\beta) &= P(1,1|\alpha,\beta) - P(1,0|\alpha,\beta) \\ &+ P(1,-1|\alpha,\beta) - P(0,1|\alpha,\beta) + P(0,0|\alpha,\beta) \\ &- P(0,-1|\alpha,\beta) + P(-1,1|\alpha,\beta) \\ &- P(-1,0|\alpha,\beta) + P(-1,-1|\alpha,\beta) \end{aligned} \quad (7)$$

Using the Bell inequality in eqn. (6) we obtain a theoretical maximum violation of 2.552 in agreement with Gisin and Peres [16]. This prediction was obtained using analyzer rotations of $\alpha = 0^o$, $\alpha' = 22.5^o$, $\beta = 11.25^o$, and $\beta' = 33.75^o$.

The entangled quanta we wish to use are the multiphoton modes of a polarization entangled field [20] produced by pulsed type-II parametric down-conversion. The first order term of parametric down-conversion is $1/\sqrt{2}\,(|H,V\rangle - |V,H\rangle)$, which is used in spin-1/2 Bell inequality experiments. A graphical representation is shown in parentheses in Fig. 1 a). However we are interested in the second order term of the down-converted field. By using postselection we can selectively measure this term, which is given by

$$\frac{1}{\sqrt{3}}\left(|2H,2V\rangle - |HV,VH\rangle + |2V,2H\rangle\right) \quad (8)$$

where the first term in the kets represent the polarization of the photons sent to Alice and the second term in the kets represent the polarization of the photons sent to Bob. For example, the $|2H,2V\rangle$ means that if Alice measures two horizontal photons, then Bob will measure two vertical photons. The photons sent to Alice have three possible measurement outcomes with equal probabilities, namely $|2H\rangle$, $|HV\rangle$ and $|2V\rangle$, which we will define as the $|1\rangle$, $|0\rangle$ and $|-1\rangle$ state respectively (see fig. 1). Thus, it is *not* the photons that are the spin-1 particles, but the two-photon polarization entangled modes.

A schematic of our experimental setup is shown in Fig. 2. The pump laser is a 120 fs pulsed, frequency doubled, Ti:Sapphire laser operating at 390 nm with an 80 MHz repetition rate. The pump enters a nonlinear beta-barium borate (BBO) crystal cut for type-II phase matching [7]. The down-converted field is then fed back into the crystal along with the retro-reflected pump beam. The difference in the round-trip path length of the pump beam and down-converted field is much smaller than the coherence length of the 5nm bandwidth frequency filtered down-converted photons. The feedback loop for the entangled fields contains a 2 mm BBO crystal rotated 90^o with respect to the optical axis of the down-conversion crystal, which compensates for the temporal walk off. Such alignment yields very good spatial and temporal overlap with which-pass interference visibilities of 98%.

The primary purpose for using the two-pass scheme is to increase the count rates. For pulsed four-photon down conversion the count rates increased by a factor of 16 for two passes as opposed to 1 pass, provided that both down-conversion fields are exactly in phase and completely indistinguishable. This leads to approximately 10 four-photon coincidence detections per second. To perform active stabilization of the phase we use the fact that under the same conditions there is maximum constructive interference for the much more intense two-photon state (singlet spin-$\frac{1}{2}$). Thus the two-photon coincidences can then act as a precision, low-noise four-photon intensity reference.

The analysis setup is shown in the dashed box in Fig. 2. Each analyzer contains a $\lambda/2$ waveplate, a polarizer, a $\lambda/4$ waveplate, a polarizing beam splitter (PBS), narrow bandwidth filters (5 nm) and 2 single photon detectors. The half-wave plates are used to set the desired α and β on Alice's and Bob's sides. The $|HV\rangle$ state is detected by having the quarter wave plate oriented 0^o with respect to the horizontal polarization axis. The photons then pass through the quarter wave plate unaltered and are split up at the PBS. The $|2H\rangle$ state is measured by inserting a

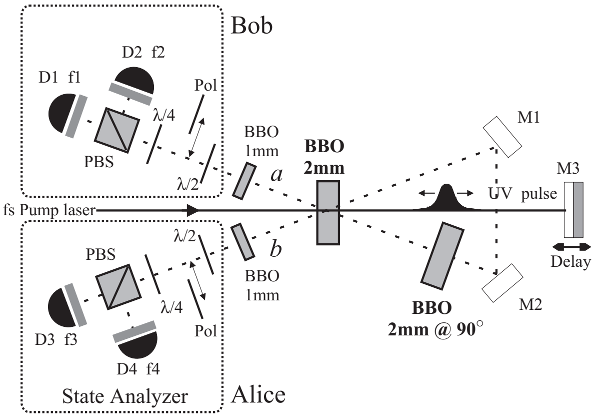

FIG. 2: Experimental setup for generation and detection of entangled spin-1 singlets. A type-II non-collinear parametric down-conversion process creates four photon states which are amplified by the double pass configuration. The detection is done at Alice's and Bob's sides by postselection as described in the main text.

linear polarizer oriented such that only horizontally polarized photons are transmitted. The quarter wave plate rotated by 45^o with the PBS is an effective 50/50 beam splitter. Thus, the probability for measuring two photons (one in each detector) on Alice's or Bob's side is reduced by a factor of 2 due to the binomial measurement statistics. In addition, inserting a polarizer introduces additional unavoidable losses in the mode and further reduces the probability of measurement compared to that of the $|HV\rangle$ state. It was experimentally determined that the two-photon measurement probability of the $|2H\rangle$ state was 43.1 % on Alice's side and 43.4 % on Bob's side compared to 50% for an ideal 50/50 beam splitter and lossless polarizer. Measuring the $|2V\rangle$ is the same as the $|2H\rangle$ except that the polarizer is rotated by 90^o.

With the configuration just described, it is necessary to measure 36 probabilities, nine from eqn. (7), for each of the four analyzer settings in eqn. (6). The experimental results for one analyzer setting (namely $\alpha = -16^o$,$\beta' = 14^o$) are listed in the table below. The measurements were taken by observing the raw 4-fold coincidence counts of all nine measurement possibilities. Each data point is the average over twelve 60 second intervals. The data obtained using two polarizers were then multiplied by a factor 1/(0.431)(0.434). The data obtained using a polarizer on Alice's (Bob's) side were multiplied by a factor of 1/(0.431) (1/(0.434)). This modified data is shown under the Mod. column and the corresponding probability under Prob. Similar tables have been measured for the other three analyzer orientations (using $\alpha' = 4^o$, $\beta = 6^o$). Combining all this data we arrive at a single value, $S = 2.27 \pm 0.02$.

$\alpha = -16^o$, $\beta' = 14^o$	$\langle Counts(60s)\rangle$	Mod.	Prob.
P(1,1)	2.20	11.71	2.25%
P(1,-1)	18.04	96.05	18.46%
P(-1,1)	17.37	92.48	17.77%
P(-1,-1)	1.78	9.48	1.82%
P(1,0)	21.92	50.47	9.70%
P(0,1)	33.67	77.86	14.96%
P(-1,0)	21.43	49.34	9.48%
P(0,-1)	28.74	66.46	12.77%
P(0,0)	66.50	66.50	12.78%
Total	-	520.35	100%

Due to the rotational symmetry of the maximally entangled state described in eqn. (8), the only relevant experimental setting to obtain a maximum violation, is the difference in angles between the analyzer settings ($\Delta\phi = \beta - \alpha = \alpha' - \beta = \beta' - \alpha'$). However, we do expect some degree of mixture in the state created in our source. A simple model for the noise in our source is given by a statistical mixture of the individual terms of the pure state. The resulting density matrix is given by

$$\rho = p\left(|\psi_{pure}\rangle\langle\psi_{pure}|\right) + \frac{(1-p)}{3}\left(|2H, 2V\rangle\langle 2H, 2V| + |HV, VH\rangle\langle HV, VH| + |2V, 2H\rangle\langle 2V, 2H|\right) \quad (9)$$

where p is the probability of having the pure entangled state. This model is a simple function of only one parameter (p), which is directly related to the lowest fringe visibility obtained by fixing the polarization orientation of one analyzer and rotating the other. This is often referred to as entanglement visibility. The presence of noise will degrade this entanglement visibility and break the rotational symmetry. Because of this symmetry breaking, it will no longer be irrelevant where we define the zero of the analyzers. In particular we want to set our measurement axes ($\alpha, \beta, \alpha', \beta'$) such that they are symmetric around the origin defined by the optical axis of the down-conversion crystal; in this way we minimize the effect of the classical noise in achieving a Bell violation. The maximum violation for a given level of noise occurs at a reduced angle difference $\Delta\phi$ compared with the ideal noiseless case [26]. The curves in fig. 3 are calculated values of S as a function of the angle difference $\Delta\phi$ for various levels of noise corresponding to the indicated entanglement visibilities. For a 100% visibility the maximum value of S is 2.55. In our experiment we measured an entanglement visibility of 75% which –in our modelling of the noise– corresponds to $p = 0.69$ and has a maximum of $S = 2.28$ for $\Delta\phi = 10$. This is in good agreement with our measured value of $S = 2.27 \pm 0.02$ at $\Delta\phi = 10^o$. This is more than 13 standard deviations away from the maximum value explainable by local realistic theories, $S = 2$. In order to rule out systematic errors we measured three additional points along the curve of 75% visibility. Each

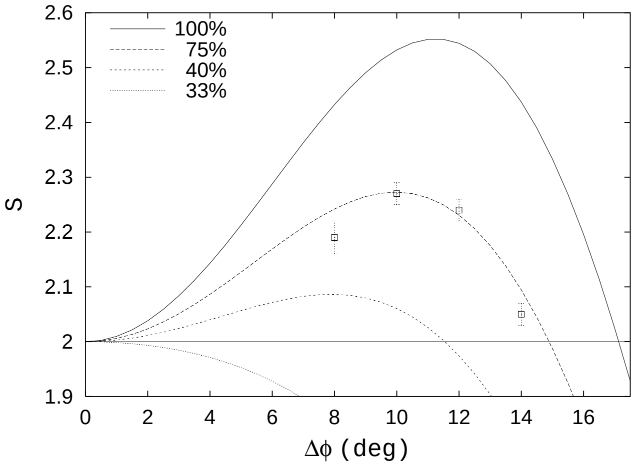

FIG. 3: The value of S is plotted as a function of the angle difference between analyzer axes. The curves correspond to different entanglement visibilities. Our experiment had 75% entanglement visibility and the experimental points are shown along with the corresponding theoretical prediction.

of these also violates the Bell inequality as expected.

Since our entangled spin-1 objects are constructed from four photons produced by parametric down-conversion, it is natural to consider whether the observed violation could be explained in terms of a product state of spin-1/2 pairs. However we have performed calculations showing that the four-photon state created in this way does not violate the inequality in 6.

Stimulated emission, which has been used to enhance our four-photon counts, also enhances Bell-type experiments by improving mode matching of the source to the detectors. We observed an increase in detection efficiency from 12% for a single pass to 18% for a double pass. With many passes through the crystal [24] and improved coupling/detection optics[25], it might be possible to obtain efficiencies high enough to perform completely loophole-free violations of Bell inequalities.

In summary, we have reported the first experimental violation of a spin-1 Bell inequality. The experimentally determined value was 2.27 ± 0.02 which is in excellent agreement with the value of 2.28 expected from the entanglement visibility of 75%. In principle, the method can be extended to higher spin numbers. These results open up the exploration of spin-1 (and higher) states for optical quantum information.

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